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# TECHNICAL NOTE

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# DELINEATION OF TRACKS OF HEAVY COSMIC RAYS AND NUCLEAR PROCESSES WITHIN LARGE SILVER CHLORIDE CRYSTALS

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SUMMARY

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Tracks of energetic charged particles, such as heavy primary cosmic rays and the products of nuclear collisions, have been made visible within the interior of large, transparent crystals of silver chloride. The tracks are delineated by photoelectric formation of metallic silver along them. This technique may be useful as a convenient and distortion-free method for the study of heavy primaries and fission fragments.

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# DELINEATION OF TRACKS OF HEAVY COSMIC RAYS AND NUCLEAR PROCESSES WITHIN LARGE SILVER CHLORIDE CRYSTALS*

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## INTRODUCTION

The study of cosmic ray particles by means of photographic emulsions has been reviewed by Powell, Fowler, and Perkins (Reference 1). Photographic emulsions, while representing a very powerful and useful technique of study, do have disadvantages such as high sensitivity to the exact processing conditions and distrotion of tracks resulting from swelling and drying of the gelatine. Thus, exploration for other recording methods seems worthwhile.

This paper describes the development of one such alternative technique–the delineation of tracks throughout the interior of single crystals of silver chloride. Crystals as large as about $1.5 \times 1.0 \times 0.5$ cm have been employed; and the use of larger crystals would also be feasible. Preliminary reports have been published elsewhere (References 2 and 3). The tracks are made visible by "decoration" with print-out silver, and have been reproduced without distortion.

It is perhaps of interest to briefly review the photographic process as normally employed, and compare it with the physical processes involved in the formation of print-out silver within a large crystal. In an emulsion, the cosmic ray particle registers its passage through the silver halide microcrystal by the liberation of electrons which ultimately combine with silver ions to form metallic specks on the surface of the microcrystal (References 4 and 5). During subsequent chemical development, these specks catalyze the reduction of the entire crystal to metallic silver. In the interior of a large crystal, however, the decomposition products (silver and a halogen) have little chance of reaching the surface before recombination; moreover, any silver specks formed in the interior cannot interact with a chemical developer applied to the surface. The registration and magnification of the trajectories of particles through large crystals must therefore rely upon other phenomena.

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The passage of a charged particle through matter produces local vibrational and electronic excitation, resulting in intense heating of a cylindrical region about the track (see Reference 6). In a crystal as plastic as silver chloride the expansion of this hot region deforms the surrounding material, and the cylinder is in turn deformed as it is rapidly quenched (within about $10^{-11}$ second) to room temperature (see References 7 and 8). The track of a charged particle is thus surrounded by a cylindrical region of severely deformed matter. It should be possible to decorate this track by applying the techniques already developed for the study of dislocations in crystals.

During the past decade there has been an extensive development of methods for making dislocations visible by the deposition of matter of some sort on them. The technique of precipitation of a solute from a supersaturated solid solution (see Reference 9) presumably could not be applied to cosmic ray track delineation, because annealing the crystal would probably decrease the damage in the track. Hedges and Mitchell (Reference 10), however, were able to decorate dislocations in AgBr at room temperature by photolyzing the regions near the crystal surface. Moreover, Haynes and Shockley (Reference 11) showed that with pulsed photoresponse techniques it is possible to sweep photoelectrons many millimeters through AgCl. Each electron trapped in the interior then gives rise to an atom of metallic silver, by virtue of the ability of interstitial silver ions to migrate to the trapping sites. A combination of these two techniques has been shown to be capable of delineating imperfections throughout large crystals (see Reference 12), and it is this process that has been employed in the present work.

## EXPERIMENTATION

The specimens employed in this work were in the form of slabs, cut from Harshaw Chemical Company AgCl single crystal discs. It was found that only those specimens for which the polyvalent metal impurity content was less than six parts per million (as gauged from the ionic conductivity) gave good decoration. The careful removal (by means of polishing papers and cloths moistened with 3% KCN solution) of 2 to 3 mm from one of the broad surfaces was necessary to produce a relatively strain-free surface through which the photoelectrons could subsequently be driven. The crystals were annealed in air on finely powdered silica for 14 hours at 425°C, and returned to room temperature at a rate of no more than 12°C/hour. They were again etched with KCN solution.

The crystals were mounted in plexiglass holders which also carried nuclear photographic emulsions. The strain-free face of each crystal was parallel to an emulsion slab, and separated from it by only a 1/4-mil sheet of Mylar. The positioning of the emulsion and crystal was well defined, so that after separation of the two it would be possible to transform from coordinates based on the emulsion to those based on the crystal.

These holders, wrapped in Myiar and in black plastic tape to produce a light-tight package, were flown in a balloon for 9 hours at an altitude of 108,000 feet. The orientation of the package during the flight was such that the emulsion slab was above the crystal, and both were inclined 30 degrees to the horizontal.

Some three months after the flight, the specimens were subjected to the internal decoration procedure; photoelectrons created at the strain-free surface by an ultraviolet mercury lamp were swept into the crystal by an electric field. Although AgCl is an ionic conductor at room temperature, the relaxation time of an internal electric field in AgCl is hundreds of microseconds, as compared with a photoelectron lifetime on the order of microseconds. Thus, if the ultraviolet radiation is applied in pulses a few microseconds in length, and spaced at about one millisecond, and if the sweeping field is pulsed synchronously with the light flashes, the photoelectrons may be displaced before the ionic conductivity can produce appreciable relaxation. Various mechanical and electronic arrangements to provide these synchronous pulses have been described by Haynes and Shockley; Webb; Hamilton, Hamm, and Brady; and Süptitz (References 11, 13, 14, and 15). The apparatus employed in this work (Reference 16) applies a potential difference of 2 kv across the crystal, and obtains the ultraviolet flashes from a General Electric BH-6 lamp at a repetition rate of 1000 per second. Approximately $10^9$ photoelectrons per $cm^2$ of crystal surface are produced in each flash.

In a crystal several millimeters thick, most of the photoelectrons are usually trapped within the crystal at the sites of imperfections or impurities. Mobile interstitial silver ions with jump frequencies of almost $10^{11}$/sec (Reference 17) migrate to trapped electrons to form metallic silver atoms. Repeated electron trapping and neutralization result in the formation of specks of silver about one micron in size. At the intensity and pulse repetition rate used in this work, an exposure of two hours is sufficient to form a colloidal distribution within the crystal. A typical crystal, as seen by scattered light, is shown in Figure 1. Observation under a conventional microscope shows that (if the material is sufficiently pure) the Ag specks delineate dislocations and the tracks of energetic charged particles.

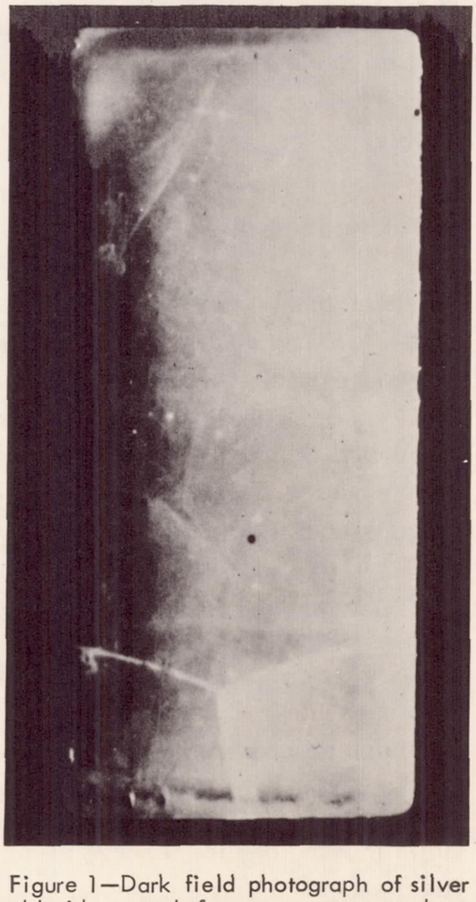

Figure 1—Dark field photograph of silver chloride crystal after exposure to synchronous electric field and ultraviolet light. The crystal is 4.8mm high and 10mm wide. The lines visible in the photograph are grain boundaries.

## OBSERVATIONS

A one-to-one correspondence between the tracks of heavy (carbon and above) primary cosmic rays in the emulsions and in the crystals was established. Every heavy particle track in an emulsion was also observed in the corresponding crystal, at the expected location and orientation. Moreover, there was a correspondence between track widths in the emulsions and widths of the same tracks in the crystals. Figure 2 shows one such track inside a crystal, and Figure 3 shows a portion of another track in the emulsion along with its continuation in the crystal. No tracks attributable to protons, alphas, or light primaries have been found in the crystals; apparently the density of energy released by particles of low charge and high velocity is too small to produce much thermal strain in the crystals. Control experiments were performed on many crystals which were not flown at high altitudes, and no patterns attributable to cosmic ray tracks were ever observed in these.

It was always possible to follow the tracks of the heavy primaries completely throughout the 5-mm thickness of the crystals, as shown in Figure 4. The apparent increase in track width in the photographs of track segments below about 3000$\mu$ is due to the scattering of light within the crystal. This was demonstrated by viewing the same track through the opposite surface of the crystal.

Only one primary track was found which ended within the crystal; a large increase in track density was apparent near the end of its range.

In addition to the straight tracks of the primary radiation, twelve events with two or more secondary particles were found. Examples of these are given in Figures 5 through 9. They were apparently induced by fast light primaries, since the tracks of the incoming particles are not visible.

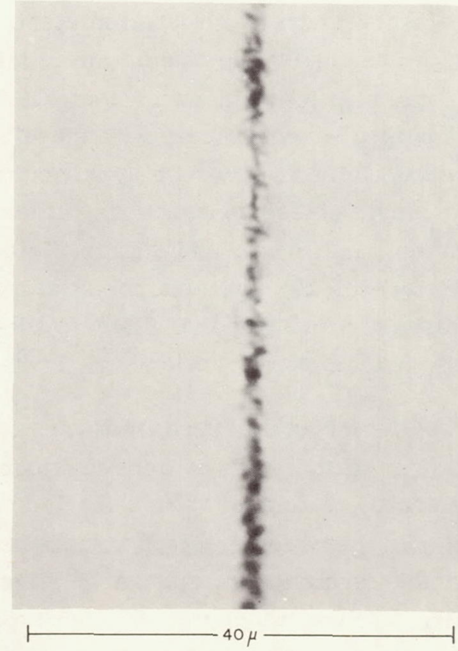


Figure 2—Heavy cosmic ray particle track inside the crystal of Figure 1.

## CONCLUSION

It is thus established that the internal print-out technique is capable of recording the tracks of energetic charged particles, provided that the energy loss density is great enough. Therefore, at relativistic speeds, only highly charged particles are seen; alpha particles are recorded if they move slowly enough to release sufficient energy to damage the crystal thermally. For example, this technique has been used to detect nuclear disintegrations produced by 1.55 Bev protons, in which more than twelve secondary particles had ranges greater than 1800 $\mu$ which is equivalent to about 3-1/2 mm of emulsion (Reference 15).

Further experiments will be necessary in order to evaluate the potential utility of this technique. The question of whether the track width in the crystal is a reliable measure of energy loss rate—and hence, of the charge of relativistic particles—must be settled. Extensive comparison of tracks in crystals and emulsions would then permit a calibration of the method. The effects of crystal purity and the possible use of AgBr as well as AgCl should

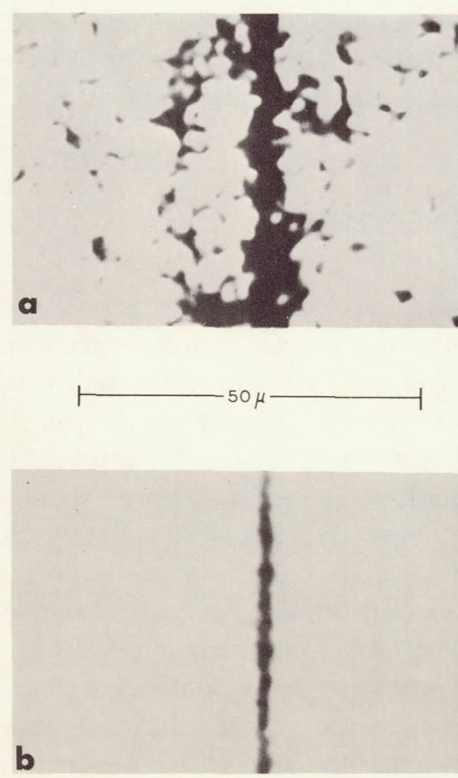


Figure 3—Tracks of the same cosmic ray particle (a) in the adjacent emulsion and (b) 72 $\mu$ below the crystal surface.

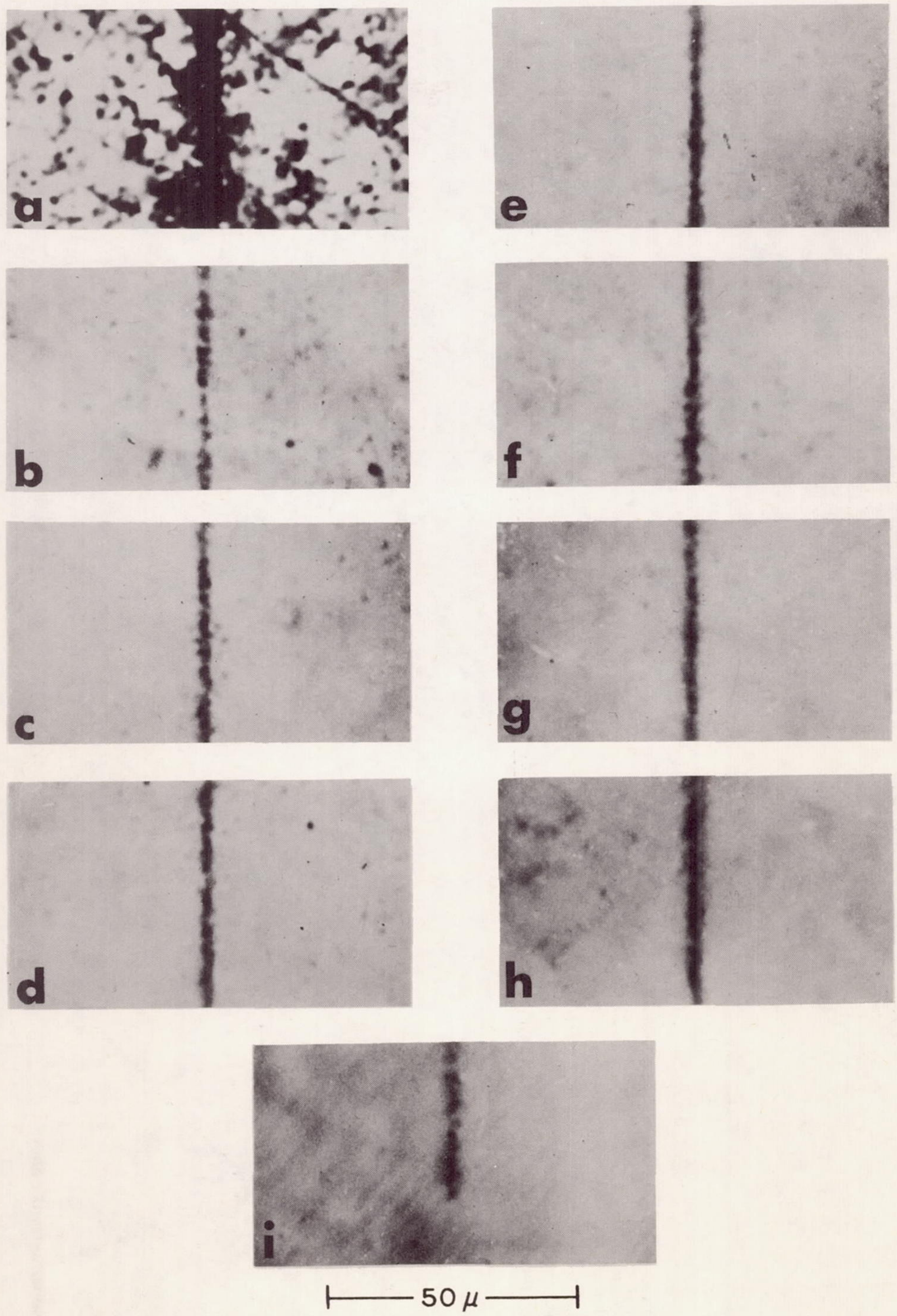


Figure 4—Cosmic ray particle track (a) in the emulsion, and within the crystal at depths of (b) 500 $\mu$; (c) 1000 $\mu$; (d) 1500 $\mu$; (e) 2000 $\mu$; (f) 2500 $\mu$; (g) 3000 $\mu$; (h) 3500 $\mu$; (i) bottom surface, 4860 $\mu$. The apparent increase in track diffuseness below about 3000 $\mu$ is due to scattering of light in the crystal.

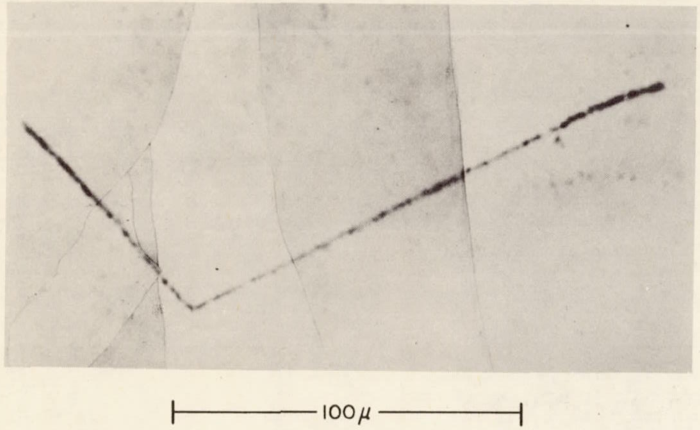


Figure 5—Two secondary particles produced in a collision at a depth of 1560μ below crystal surface. Note the increase in the track densities near the ends of the ranges. This Figure and those following are composites of several photographs taken at different depths. The faint, irregular lines in the Figures are the edges of the individual photographs.

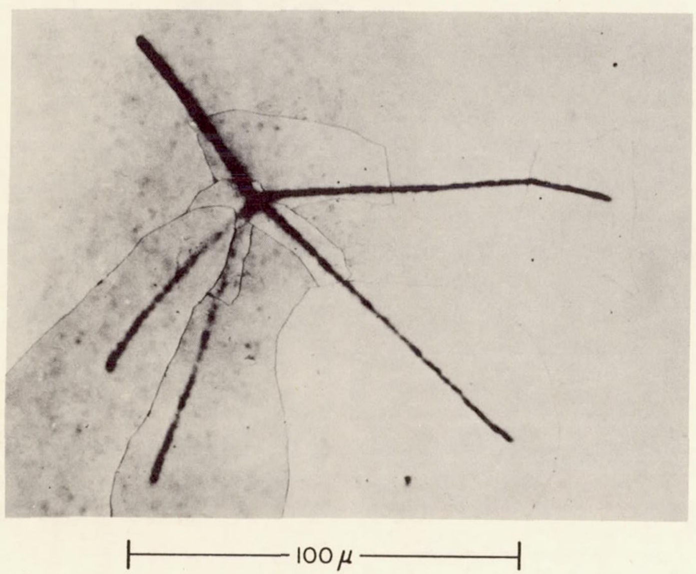


Figure 6—A five-prong star produced 1440μ below the crystal surface.

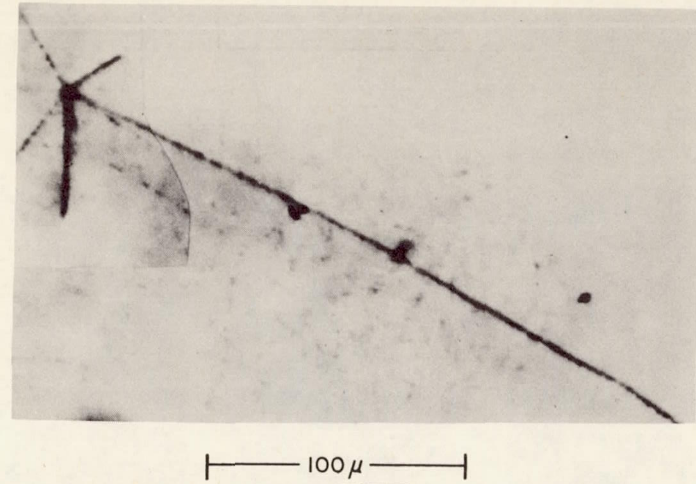


Figure 7—A star with eight prongs, two of which are only lightly decorated, produced 1500 μ below the crystal surface.

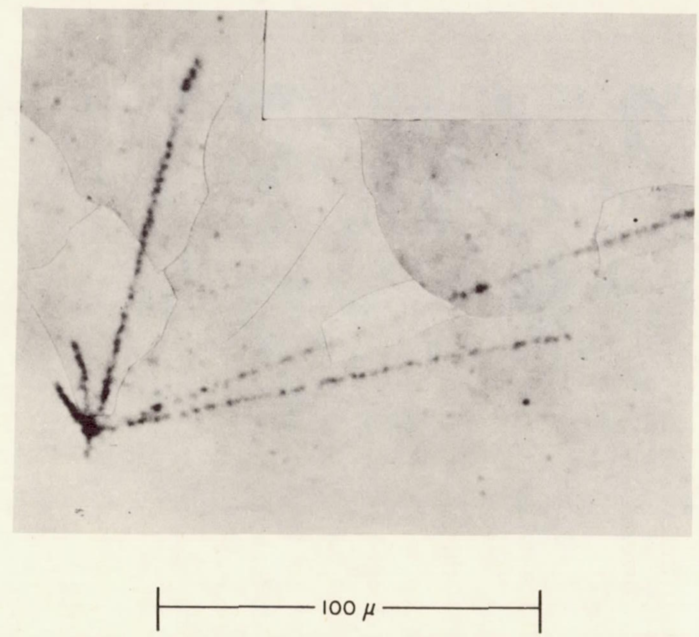


Figure 8—A six-prong star observed at a depth of 1500μ below crystal surface.

be investigated. Finally, elimination of the dislocation background by the recrystallization method of Bartlett and Mitchell (Reference 19) seems promising. These investigations are currently in progress.

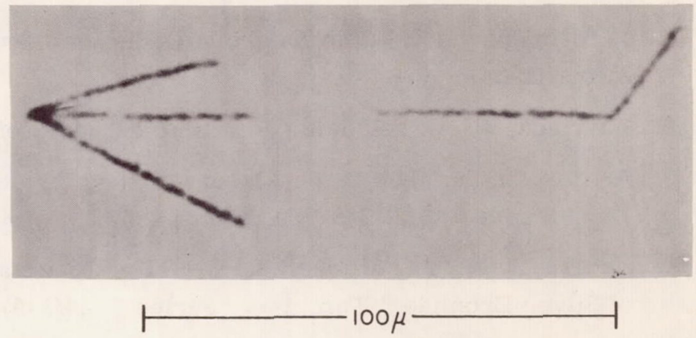


Figure 9—A three-prong star produced at 1740$\mu$ below the crystal surface. A poorly decorated segment, such as that seen in the center of the long track of this figure, is quite uncommon; the reason for this lack of decoration is not understood at present.

If the method does indeed prove to be reliable, several specialized applications suggest themselves. The collection of statistics on heavy primaries by means of prolonged exposures from recoverable satellites would now be simplified, and without the fogging normally produced by the high incidence of lighter particles. Heavy particles in solar flares could similarly be studied. Moreover, in any collision recorded in the crystal, angles could be measured rather accurately, since the development process does not distort the medium. Finally, this technique seems well-suited to the study of fission fragments, either by exposure of pure crystals to particles from high-energy accelerators (References 18 and 19), or by exposure to slow neutrons of crystals doped with fissionable material.

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